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(19) (CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) Electroluminescent Devices

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(57) 6 Claims

Notice: This application is as filed and may therefore contain an incomplete specification.



Electroluminescent devices

Abstract

An electroluminescent device which contains at least 2 electrodes, the base electrode and the top electrode, a light-emitting layer, and a stabiliser which is effective against atmospheric oxidation, thermal oxidation, ozone and UV radiation, wherein the electroluminescent device may contain at least one further layer selected from the sequence comprising the hole-injecting layer, the hole-transporting layer, and the electron-injecting layer.

Electroluminescent devices

5 An electroluminescent (EL) device is characterised in that it emits light with a flow of current when an electrical voltage is applied. Devices of this type have long been known in the art under the description "light-emitting diodes" (LEDs = light emitting diodes). The emission of light occurs due to the recombination of positive charges ("holes") and negative charges ("electrons") with the radiation of light.

10 Inorganic semiconductors such as gallium arsenide are mainly used nowadays for the development of light-emitting components for electronics or photo-electronics. Point source display elements can be produced based on substances of this type. Large-surface devices are not possible.

15 In addition to semiconductor light-emitting diodes, electroluminescent devices based on vapour-deposited low molecular weight organic compounds are known (US-P 4 539 507, US-P 4 769 262, US-P 5 077 142, EP-A 406 762). Only LEDs of small dimensions can be produced with these materials also - due to the production process. Moreover, these electroluminescent devices only have very short lifetimes.

20 Polymers such as poly-(p-phenylenes) and poly-(p-phenylenevinylenes (PPV)) have also been described as electroluminescent polymers: G. Leising et al., Adv. Mater. 4 (1992) No. 1; Friend et al., J. Chem. Soc., Chem. Commun. 32 (1992); Saito et al., Polymer, 1990, Vol. 31, 1137; Friend et al., Physical Review B, Vol. 42, No. 18, 11670, or WO 90/13148. Other examples of PPV in
25 electroluminescent displays are described in EP-A 443 861, and in WO-A-9203490 and 92003491.

EP-A 0 294 061 discloses an optical modulator based on polyacetylene.

Heeger et al. have proposed soluble, conjugated PPV derivatives for the production of flexible polymer LEDs (WO 92/16023).

30 Organic EL devices generally contain one or more layers of organic charge-transport compounds. The basic structure within the sequence of layers is as follows:

- 1 support, substrate
- 2 base electrode
- 3 hole-injecting layer
- 4 hole-transporting layer
- 5 5 light-emitting layer
- 6 electron-transporting layer
- 7 electron-injecting layer
- 8 top electrode
- 9 contacts
- 10 10 cladding, encapsulation.

This structure constitutes the most general case, and can be simplified by omitting individual layers so that one layer assumes a plurality of functions. In the simplest case, an EL device consists of two electrodes, between which there is an organic layer which performs all the functions - including that of the emission of light. Systems of this type based on poly(p-phenylene vinylene) are described in the Application WO 90/13148, for example.

A common feature of all these EL devices which are based on low molecular weight compounds is that they do not have a satisfactory operating lifetime. During operation, organic light-emitting diodes become very hot ($> 100^{\circ}\text{C}$), and this results in a change in the layers (up to their destruction), so that a reduction of their performance or a complete loss of function then occurs. In these LED displays based on organic materials, the long-term stability of the luminous layer is a problem.

It has now been found that the long-term stability of the electroluminescent device can be significantly increased by additive substances.

The present invention relates to electroluminescent devices which comprises at least 2 electrodes, the base electrode 2 and the top electrode 8, a light-emitting layer 5, and a stabiliser which is effective against atmospheric oxidation, thermal oxidation, ozone and UV radiation, wherein the electroluminescent device may contain at least one further layer selected from the sequence comprising the hole-injecting layer 3, the hole-transporting layer 6, and the electron-injecting layer 7.

The stabilisers may be added to one or more of layers 3-7. The stabilisers are preferably introduced into the light-emitting layer 5.

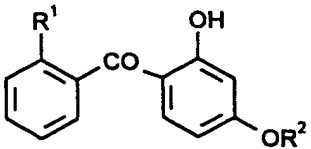
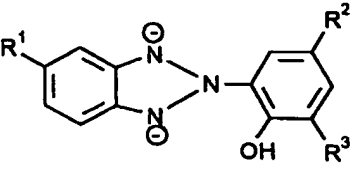
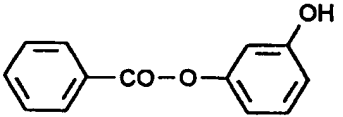
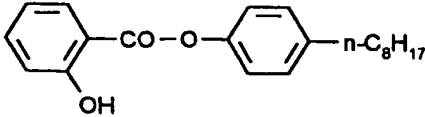
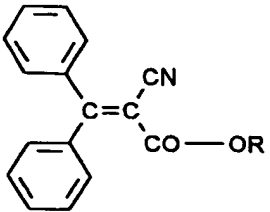
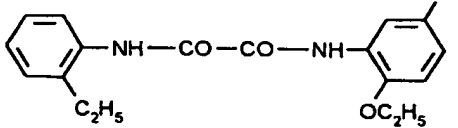
5 The amounts of stabilisers added are between 0.1 and 10 % by weight, with respect to the weight of the layers to which the stabilisers are added. 0.2 to 3 % by weight of stabiliser is preferably used.

The stabilisers are usually deposited from solution, together with the remaining layer constituents of layers 3-7. The stabilisers may also be deposited on their own as an additional intermediate layer from solution or may be vacuum-deposited as a pure compound.

10 In the case of UV absorbers, the stabilisers may also be deposited as an uppermost additional layer on the support or substrate 1. For this purpose, the UV absorbers are used in admixture with a binder, such as polystyrene, polyurethane, polyvinyl acetate, polyacrylate, polycarbonate, polyolefines or polysulphone, which facilitates bonding to 1. High stabilities are preferably obtained by using the stabilisers
15 in crosslinking systems such as those described in EP-A-637 899.

Examples of suitable stabilisers include UV stabilisers such as those listed in Table 1 (the number of the compound is given in brackets in each case).

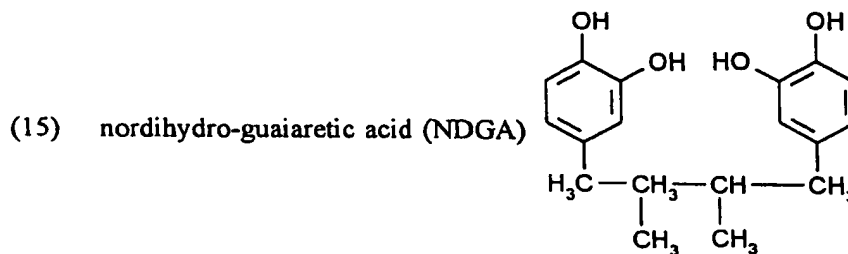
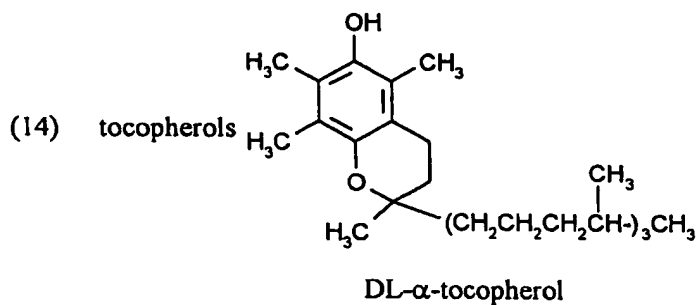
Table 1

5	(1)	$R^1 = H, R^2 = CH_3$	
	(2)	$R^1 = H, R^2 = n-C_8H_{17}$	
	(3)	$R^1 = OH, R^2 = CH_3$	
	(4)	$R^1 = H, R^2 = n-C_{12}H_{25}$	
	(5)	$R^1 = COOH, R^2 = CH_3$	
10	(6)	$R^1 = H, R^2 = CH_3, R^3 = H$	
	(7)	$R^1 = H, R^2 = \text{tert.-butyl}, R^3 = \text{tert.-butyl}$	
	(8)	$R^1 = Cl, R^2 = CH_3, R^3 = \text{tert.-butyl}$	
	(9)	$R^1 = H, R^2 = \text{tert.-butyl}, R^3 = \text{tert.-butyl}$	
	(10)	$R^1 = Cl, R^2 = \text{tert.-amyl}, R^3 = \text{tert.-amyl}$	
15	(11)		
	(12)		
	(13)	$R = C_2H_5$	
	(14)	$R = CH_2-CH_n-C_4H_9$ $ $ C_2H_5	
	(15)	$R = \text{tert.-butyl}$	
	(16)	$R = H$	

In addition, antioxidants and light stabilisers, e.g. those based on sterically hindered phenols, may be used as stabilisers (see Table 2).

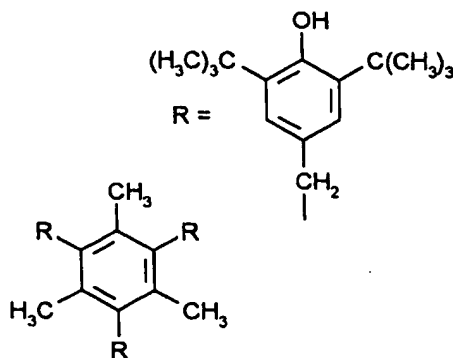
Table 2

- | | | |
|----|------|---|
| | (1) | 2,6-di-tert.-butyl-phenol |
| 5 | (2) | 2,6-di-tert.-butyl-4-methyl-phenol |
| | (3) | 2,4,6-tri-tert.-butyl-phenol |
| | (4) | 2,6-di-tert.-butyl-4-nonyl-phenol |
| | (5) | 6-tert.-butyl-2,4-dimethyl-phenol |
| | (6) | 2,4-dimethyl-6-nonyl-phenol |
| 10 | (7) | 2,4-dimethyl-6-(1-phenyl-ethyl)-phenol |
| | (8) | 2,4-dimethyl-6-(1-methyl-cyclohexyl)-phenol |
| | (9) | 2,6-di-octadecyl-4-methyl-phenol |
| | (10) | (5-tert.-butyl-4-hydroxy-3-methyl-benzyl)-malonic acid-di-n-octadecyl ester |
| | (11) | 2,6-di-tert.-butyl-4-methoxy-phenol, 3,5-di-tert.-butyl-4-hydroxy-anisole |
| 15 | (12) | 2,5-di-tert.-butyl-hydroquinone (DBH) |
| | (13) | 2,5-bis-(1,1-dimethyl-propyl)-hydroquinone |

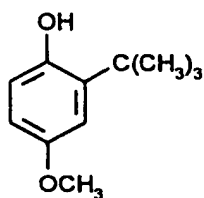


- | | | |
|----|------|---|
| 20 | (16) | α- and β-naphthol |
| | (17) | 6,7-dihydroxy-4-methyl-coumarin |
| | (18) | 5,7-dihydroxy-4-methyl-coumarin monohydrate |
| | (19) | 1,3,5-trihydroxy-benzene, phloroglucinol |

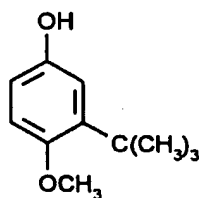
- (20) 3,4,5-tri-hydroxy-benzoic acid propyl ester = propyl gallate, PG
 (21) 3,4,5-tri-hydroxy-benzoic acid octyl ester = octyl gallate, OG
 (22) 3,4,5-tri-hydroxy-benzoic acid dodecyl ester = dodecyl gallate, lauryl gallate, LG
 5 (23) 2,4,5-trihydroxy-butyrophenone = THBP
 (24) 2,2'-methylene-bis-(4-ethyl-6-tert.-butyl-phenol)
 (25) 1,1-bis-(2-hydroxy-3,5-dimethyl-phenyl)-butane
 (26) 1,1'-methylene-bis-(naphthol-2)
 (27) 2,2-bis-(4-hydroxy-phenyl)-propane = bisphenol A
 10 (28) mixture of tert.-butylated 2,2-bis-(4-hydroxy-phenyl)-propanes
 (29) bis-3,3-bis-(4-hydroxy-3-tert.-butyl-phenyl)-butanoic acid glycol ester = DTB glycol ester
 (30) 1,1-bis-(5-tert.-butyl-4-hydroxy-2-methyl-phenyl)-butane
 (31) 1,1,3-tris-(5-tert.-butyl-4-hydroxy-2-methyl-phenyl)-butane
 15 (32) 4,4'-methylene-bis-(2-tert.-butyl-6-methyl-phenol)
 (33) 4,4'-methylene-bis-(2,6-di-tert.-butyl-phenol)
 (34) 4,4'-methylene-bis-(2,5-di-tert.-butyl-phenol)
 (35) 1,1-bis-(4-hydroxy-phenyl)-cyclohexane
 (36) 1,1-bis-(3-cyclohexyl-4-hydroxy-phenyl)-cyclohexane
 20 (37) 1,3,5-trimethyl-2,4,6-tris-(3,5-di-tert.-butyl-4-hydroxy-benzyl)-benzene:



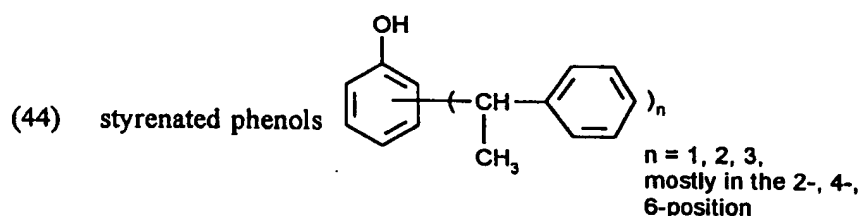
- (38) catechol
 (39) 4-tert.-butyl-catechol = TBC; 1,2-dihydroxy-4-tert.-butylbenzene
 (40) hydroquinone
 25 (41) 4-methoxyphenol, hydroquinone monomethyl ether
 (42) 4-benzyloxyphenol, hydroquinone monobenzyl ether
 (43) mixtures of 3-tert.-butyl-4-hydroxy-anisole



3-isomer



2-isomer

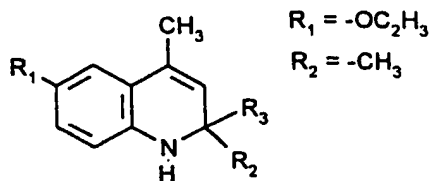


- (45) 3,5-di-tert.-butyl-4-hydroxy-benzyl alcohol
 (46) 2,6-di-tert.-butyl-4-methoxy-phenol
 5 (47) octadecyl-3-(3,5-di-tert.-butyl-4-hydroxy-phenyl)-propionate
 (48) pentaerythrityl-tetrakis-[3-(3,5-di-tert.-butyl-4-hydroxy-phenyl)-propionate]
 (49) 1,6-bis-[3-(3,5-di-tert.-butyl-4-hydroxy-phenyl)-propionyloxy]-n-hexane
 (50) 2,2-bis-(3,5-di-tert.-butyl-4-hydroxy-benzyl)-malonic acid di-n-octyl ester
 (51) 2,2'-methylene-bis-(4,6-dimethyl-phenol)
 10 (52) 2,2'-methylene-bis-(6-tert.-butyl-4-methyl-phenol)
 $R = -C(CH_3)_3$
 (53) 2,2'-methylene-bis-(4-methyl-6-nonyl-phenol)
 (54) 2,2'-methylene-bis-[4-methyl-6-(1-methyl-cyclohexyl)-phenol]
 (55) 2,2'-methylene-bis[4-methyl-(6- α -methyl-benzyl)-phenol]
- 15 Furthermore, antiozonants can be used as stabilisers, e.g. those from the benzo-furan series, those from the enol series, those based on phenylenediamines, such as N-phenyl-1 or 2-naphthylamine, N,N'-diisopropyl-p-phenylenediamine, N,N'-di-sec.-butyl-p-phenylenediamine, or N,N'-bis-(1-ethyl-3-methylpentyl)-p-phenylene-diamine. Examples of these are listed in Table 3:

20 **Table 3:**

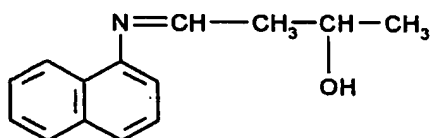
- (1) N,N'-di-sec.-butyl-p-phenylenediamine = DBPPD
 (2) N,N'-bis-(1,4-dimethyl-pentyl)-p-phenylenediamine

- (3) N,N'-bis-(1-ethyl-3-methyl-pentyl)-p-phenylenediamine
- (4) N,N'-bis-(1-methyl-heptyl)-p-phenylenediamine
- (5) N,N'-dicyclohexyl-p-phenylenediamine
- (6) N,N'-diphenyl-p-phenylenediamine = DPPD
- 5 (7) N,N'-di-(naphthyl-2)-p-phenylenediamine
- (8) N-isopropyl-N'-phenyl-p-phenylenediamine
- (9) N-(1,3-dimethyl-butyl)-N'-phenyl-p-phenylenediamine
- (10) N-(1-methyl-heptyl)-N'-phenyl-p-phenylenediamine
- (11) N-cyclohexyl-N'-phenyl-p-phenylenediamine
- 10 (12) 4-(p-toluene-sulphonamido)-diphenylamine
- (13) N,N'-dimethyl-N,N'-di-sec.-butyl-p-phenylenediamine
- (14) diphenylamine
- (15) 4-isopropoxy-diphenylamine
- (16) N-phenyl-1-naphthylamine
- 15 (17) N-phenyl-2-naphthylamine
- (18) octylated diphenylamine, predominantly 4-octyl-diphenylamine
- (19) 4-n-butylamino-phenol
- (20) 4-butyrylamino-phenol
- (21) 4-nonanoylamino-phenol
- 20 (22) 4-dodecanoylamino-phenol
- (23) 4-octadecanoyl-amino-phenol
- (24) di-(4-methoxy-phenyl)-amine
- (25) 2,6-di-tert.-butyl-4-dimethylamino-methyl-phenol
- (26) 2,4'-diamino-diphenylmethane
- 25 (27) 4,4'-diamino-diphenylmethane
- (28) N,N,N',N'-tetramethyl-4,4'-diamino-diphenylmethane
- (29) 1,2-di-(phenylamino)-ethane
- (30) 1,2-di-[(2-methyl-phenyl)-amino]-ethane
- (31) 1,3-di-(phenylamino)-propane
- 30 (32) (o-tolyl)-biguanide
- (33) the condensation product of aniline and acetaldehyde,
- (34) aniline-aldol-condensate
- (35) the product from aniline and butyraldehyde
- (36) (polymeric) 2,2,4-trimethyl-1,2-dihydroquinoline
- 35 (37) aniline-acetone condensate
- (38) 6-ethoxy-2,2,4-trimethyl-1,3-dihydroquinoline = ethoxyquin

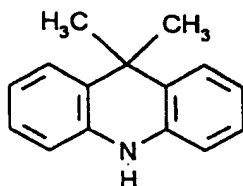


- (39) 6-dodecyl-2,2,4-trimethyl-1,2-dihydroquinoline
 (40) 2,2,4-trimethyl-6-phenyl-1,2-dihydroquinoline
 (41) 1-aminonaphthalene-aldol condensate

5



- (42) the product of 2-phenylamino-naphthalene and acetone
 (43) diphenylamine-acetone condensate, compounds of 5,5-dimethyl-acridine, etc.



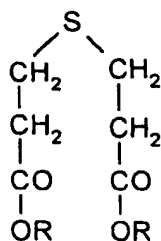
- 10 (44) N,N'-diisopropyl-p-phenylenediamine

Sulphur compounds are also suitable as stabilisers. Examples are given in Table 4.

Table 4:

- (1) 3,3'-thiodipropionic acid
 (2) 3,3'-thio-bis-(propionic acid dodecyl ester), dilauryl dithiopropionate = DLTDP

15



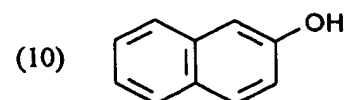
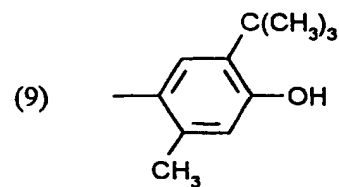
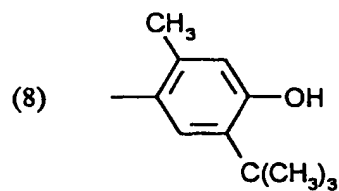
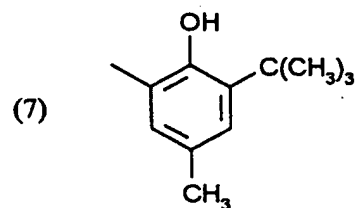
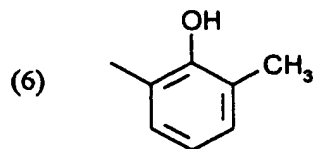
where R =

(3) $-C_{12}H_{25}$

(4) $-C_{14}H_{29}$

(5) $-C_{16}H_{27}$

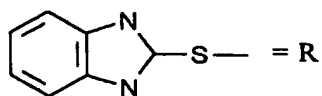
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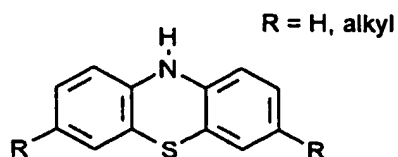
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(11) R = H

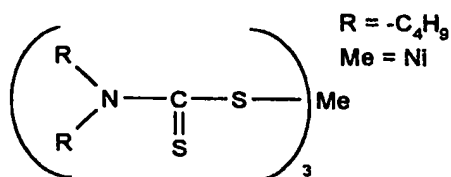
(12) R-Zn-R



(13) phenothiazine and its alkyl derivatives



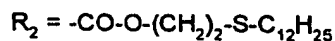
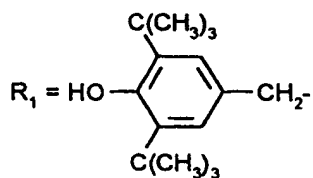
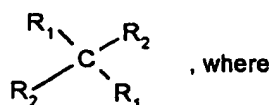
(14) nickel-(N,N'-dibutyl-dithiocarbamate)



(15) N,N'-diethylthiourea

5 (16) N,N'-dibutylthiourea

(17) bis-(3,5-di-tert.-butyl-4-hydroxybenzyl)-malonic acid-bis-(3-thiapentadecyl)
ester



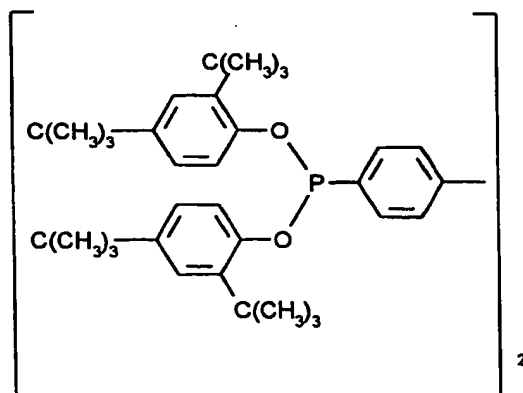
Phosphorus compounds are also suitable as stabilisers. Examples are given in
Table 5:

Table 5:

- (1) triphenylphosphine
 (2) diethyl phosphite
 (3) triphenyl phosphite
 5 (4) tris-nonylphenyl phosphite
 (5) tris-(mono-dinonylphenyl)-phosphite
 (6) tridecyl phosphite
 (7) tri-isodecyl phosphite
 (8) tri-dodecyl phosphite

10

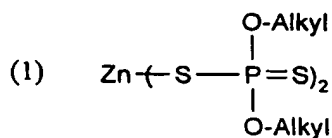
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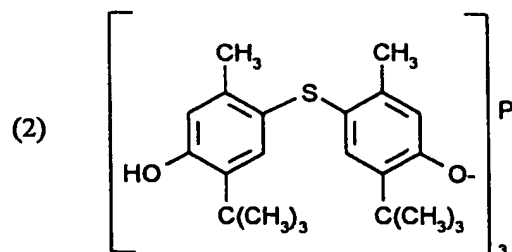


- (10) the condensation product of 4,4'-thio-bis-(2-tert.-butyl-5-methyl-phenol)
 (11) octyl-diphenyl phosphite
 (12) isooctyl-diphenyl phosphite
 (13) decyl-diphenyl phosphite
 15 (14) isodecyl-diphenyl phosphite
 (15) didecyl-phenyl phosphite
 (16) diisodecyl-phenyl phosphite
 (17) 3,5-di-tert.-butyl-4-hydroxybenzyl-phosphonic acid diethyl ester
 (18) 3,5-di-tert.-butyl-4-hydroxybenzyl-phosphonic acid di-n-octadecyl ester

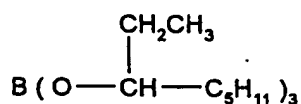
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Examples of various zinc, barium and calcium thiophosphates include

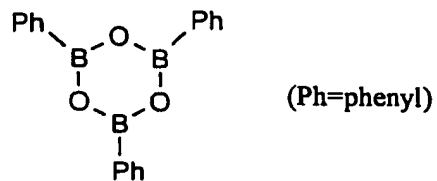
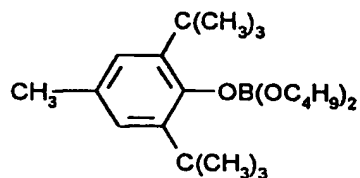
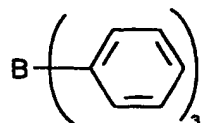




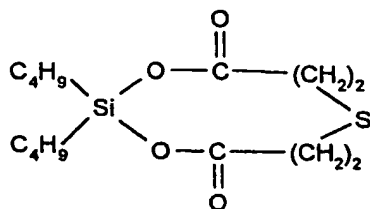
Examples of further stabilisers may comprise boric acid esters and other organo-boron compounds, e.g.

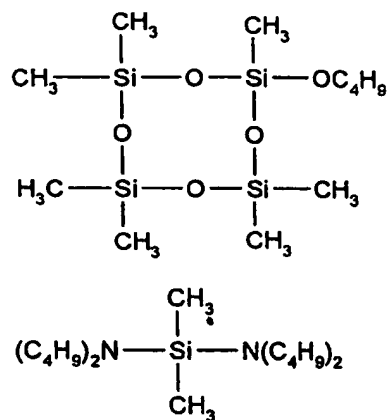


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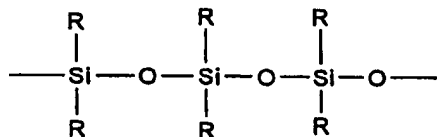


Low molecular weight silicon compounds may also be used, such as

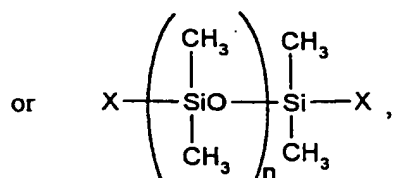




or organopolysiloxanes such as



5 where R = -CH₃, -Ph, -H oder $-(\text{CH}_2)_3\text{O}-\text{CH}_2-\text{CH}-\text{CH}_2$

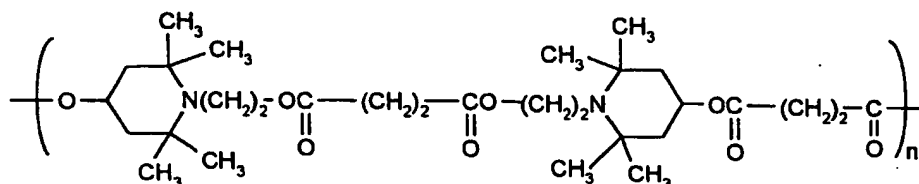
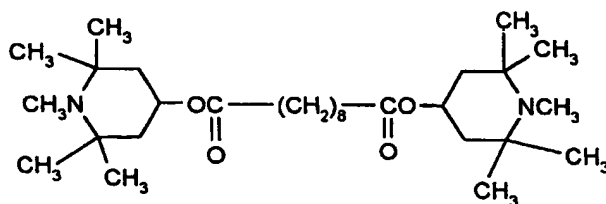
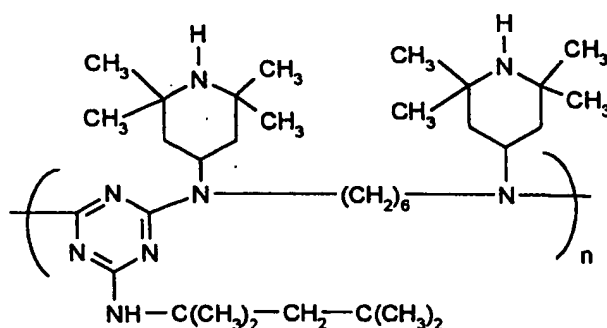


where $n = 1-10$ and

X = $-\text{CH}_2\text{O}-(\text{CH}_2)_3-\text{O}(\text{CH}_2)_3-\text{OH}$
 $-(\text{CH}_2)_4-\text{OH}$
 $-\text{OC}_2\text{H}_5$.

10

Sterically hindered amines may also be used, e.g. those based on 2,6-tetramethyl-piperidine, such as



UV absorbers, sterically hindered phenols or sterically hindered amines are preferably used as stabilisers.

Charge transport compounds according to the invention are understood to be all compounds which transport charges (holes and/or electrons) in any manner. These also expressly comprise those compounds which are constituents of the emitter

layer and which therefore constitute photoluminescent materials, such as fluorescent dyes, for example.

5 A multiplicity of organic compounds which transport charges (holes and/or electrons) is described in the literature. Low molecular weight substances which are vapour-deposited under high vacuum, for example, are predominantly used. These are mostly tertiary aryl amines or compounds containing oxadiazole groups. A good survey of these classes of substances and their use is given in the publications EP-A-387 715, and in US-P 4 539 507, 4 720 432 and 4 769 292. In principle, all substances can be used which are known as photoconductors from
10 electrophotography.

Glass or plastics material, e.g. a sheet of polyethylene terephthalate, is used as a transparent support.

The following are examples of suitable electrode materials:

- a) metal oxides, e.g. indium-tin oxide (ITO), tin oxide (NESA), etc.
- 15 b) semi-transparent metal films, e.g. Au, Pt, Ag, Cu, etc.
- c) conductive polymer films such as polyanilines, polythiophenes, etc.

Metal oxide and semi-transparent metal film electrodes are deposited as a thin layer by techniques such as vacuum-metallising, sputtering, platinising, etc.

One of the electrodes is generally transparent in the visible region of the spectrum.

20 The thickness of the transparent electrode is 50 Å to about several µm, preferably 100 Å to 5000 Å.

The electroluminescent substance is deposited directly as a thin film on the transparent electrode or on a charge-transporting layer which may optionally be present. The thickness of the film is 30 Å to 10 µm, preferably 50 Å to 1 µm.

25 After drying the EL layer, it is provided with a counterelectrode. The latter consists of a conductive substance which may be transparent.

The device according to the invention is placed in contact with the two electrodes by means of two electrical leads (e.g. metal wires).

5 When a DC voltage within the range up to 100 volts is applied, the devices emit light with a wavelength of 400 to 700 nm. They exhibit photoluminescence within the range from 400 to 700 nm.

10 The electroluminescent layer contains photoluminescent materials such as those which are described in EP-443 861, WO 9203 490 and WO 92 003 491, for example. They may also optionally contain customary additives such as inert binders, charge-transporting substances, or mixtures of inert binders and charge-transporting substances. Charge-transporting substances increase the intensity of electroluminescence and reduce the operating voltages.

15 Soluble transparent polymers are preferably used as inert binders, such as polycarbonates, polystyrene and copolymers of polystyrene such as SAN, polysulphones, polyacrylates, polyvinyl carbazole, vinyl acetate and vinyl alcohol copolymers, and copolymeric polyolefines, etc., for example. These inert binders are also suitable as binders for the stabilisers.

Example

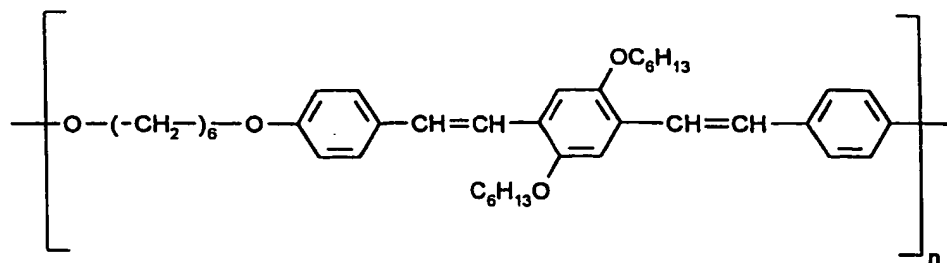
Production of electroluminescent devices of polymers, with the addition of additives to increase their storage- and operational stability.

5 ITO-coated glass (manufactured by Balzers) was cut into substrates of size 20 x 30 mm² and cleaned. In the course of this procedure, the following steps were performed in succession:

1. rinsing for 15 minutes with distilled water and falterol in an ultrasonic bath,
- 10 2. rinsing for 2 x 15 minutes with fresh distilled water each time in an ultrasonic bath,
3. rinsing for 15 minutes with ethanol in an ultrasonic bath,
4. rinsing for 2 x 15 minutes with fresh acetone each time in an ultrasonic bath,
5. drying on lint-free lens cloths.

15 A 1 % solution in dichloroethane was prepared of the polyether of formula A. 0.5 % by weight, with respect to the amount of electroluminescent polymer, of the additive TINUVIN 622LD (Ciba-Geigy) was added to the solution. The solution was then filtered (0.2 µm filter, supplied by Sartorius). The filtered solution was distributed on the ITO layer using a lacquer centrifuge operating at 100 rpm. The
20 thickness of the dry film was 120 nm. The Ra value of the surface was 5 nm (Alpha-Step 200 stylus profilometer supplied by Tencor Inst.). In parallel with this, a control specimen was produced without the additive. The polymeric layers produced in this manner were then vacuum-metallised with Al electrodes. For this purpose, isolated Al spots with a diameter of 3 mm were vacuum-evaporated on to
25 the polymer surface with the aid of a perforated mask. During the vacuum-evaporation process, the prevailing pressure in the vacuum-evaporation apparatus (Leybold) was less than 10⁻⁵ mbar.

Polyether of formula A:



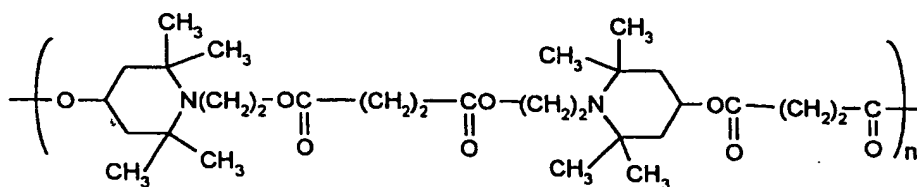
- 5 The ITO layer and the Al electrode were connected via electrical leads to a voltage source. When the voltage was increased, a current flows through the polymer layer. Electroluminescence was detectable above a voltage of 5 volts. The colour of the electroluminescence was blue-green.

Comparison of the electroluminescent devices:

dark storage period before use: 14 days

applied voltage: 17 volts.

- 10 TINUVIN 622LD:



Device structure	After 1000 seconds of operation			After 2000 seconds of operation		
	Current (mA)	EL intensity (arbitrary units)	Efficiency (arbitrary units)	Current (mA)	EL intensity (arbitrary units)	Efficiency (arbitrary units)
ITO/polyether A/Al (control specimen) without stabiliser	50	5.00E-10	1E-11	50	4.00E-10	8E-12
ITO/polyether A + 0.5 % TINUVIN 622LD/Al	2	6.00E-08	3E-08	2.2	8.00E-08	4E-08

Al = aluminium

Claims

1. An electroluminescent device which contains at least 2 electrodes, the base electrode and the top electrode, a light-emitting layer, and a stabiliser which is effective against atmospheric oxidation, thermal oxidation, ozone and UV radiation, wherein the electroluminescent device may contain at least one further layer selected from the sequence comprising the hole-injecting layer, the hole-transporting layer, and the electron-injecting layer.
2. An electroluminescent device according to claim 1, wherein the stabiliser is introduced into the light-emitting layer.
3. An electroluminescent device according to claim 1, wherein the stabiliser is added in amounts of 0.1 to 10 % by weight with respect to the weight of the layers to which the stabiliser is added.
4. An electroluminescent device according to claim 3, wherein the stabiliser is added in amounts of 0.2 to 3 % by weight.
5. An electroluminescent device according to claim 1, wherein the stabiliser is selected from the series comprising UV stabilisers, antioxidants and light stabilisers, antiozonants, sulphur compounds, phosphorus compounds, zinc, barium and calcium thiophosphates, boric acid esters, low molecular weight silicon compounds, organopolysiloxanes and sterically hindered amines.
6. An electroluminescent device according to claim 5, wherein the following stabilisers are used:

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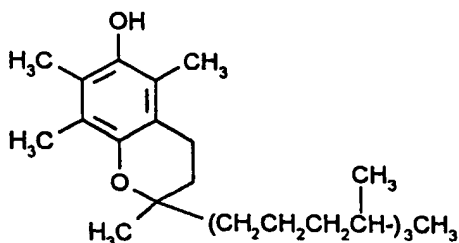
Table 1

5	(1)	$R^1 = H, R^2 = CH_3$	
	(2)	$R^1 = H, R^2 = n-C_8H_{17}$	
	(3)	$R^1 = OH, R^2 = CH_3$	
	(4)	$R^1 = H, R^2 = n-C_{12}H_{25}$	
	(5)	$R^1 = COOH, R^2 = CH_3$	
10	(6)	$R^1 = H, R^2 = CH_3, R^3 = H$	
	(7)	$R^1 = H, R^2 = \text{tert.-butyl}, R^3 = \text{tert.-butyl}$	
	(8)	$R^1 = Cl, R^2 = CH_3, R^3 = \text{tert.-butyl}$	
	(9)	$R^1 = H, R^2 = \text{tert.-butyl}, R^3 = \text{tert.-butyl}$	
		$R^1 = Cl, R^2 = \text{tert.-amyl}, R^3 = \text{tert.-amyl}$	
	(11)		
	(12)		
15	(13)	$R = C_2H_5$	
	(14)	$R = CH_2-CH_n-C_4H_9$ $\quad\quad\quad $ $\quad\quad\quad C_2H_5$	
	(15)	$R = \text{tert.-butyl}$	
	(16)	$R = H$	

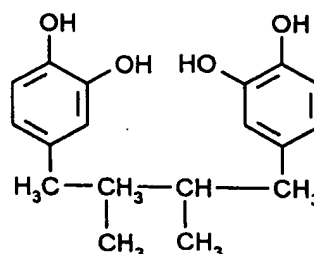
Table 2

- (1) 2,6-di-tert.-butyl-phenol
 (2) 2,6-di-tert.-butyl-4-methyl-phenol
 (3) 2,4,6-tri-tert.-butyl-phenol
 5 (4) 2,6-di-tert.-butyl-4-nonyl-phenol
 (5) 6-tert.-butyl-2,4-dimethyl-phenol
 (6) 2,4-dimethyl-6-nonyl-phenol
 (7) 2,4-dimethyl-6-(1-phenyl-ethyl)-phenol
 (8) 2,4-dimethyl-6-(1-methyl-cyclohexyl)-phenol
 10 (9) 2,6-di-octadecyl-4-methyl-phenol
 (10) (5-tert.-butyl-4-hydroxy-3-methyl-benzyl)-malonic acid-di-n-octadecyl ester
 (11) 2,6-di-tert.-butyl-4-methoxy-phenol, 3,5-di-tert.-butyl-4-hydroxy-anisole
 (12) 2,5-di-tert.-butyl-hydroquinone (DBH)
 (13) 2,5-bis-(1,1-dimethyl-propyl)-hydroquinone

15 (14) tocopherols

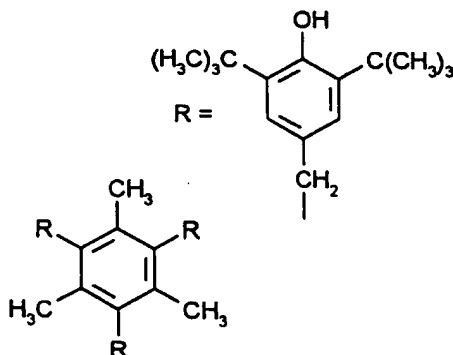
DL- α -tocopherol

(15) nordihydro-guaiaretic acid (NDGA)

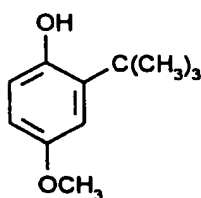


- (16) α - and β -naphthol
 (17) 6,7-dihydroxy-4-methyl-coumarin
 20 (18) 5,7-dihydroxy-4-methyl-coumarin monohydrate
 (19) 1,3,5-trihydroxy-benzene, phloroglucinol
 (20) 3,4,5-tri-hydroxy-benzoic acid propyl ester = propyl gallate, PG
 (21) 3,4,5-tri-hydroxy-benzoic acid octyl ester = octyl gallate, OG

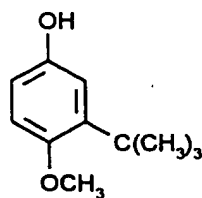
- (22) 3,4,5-tri-hydroxy-benzoic acid dodecyl ester = dodecyl gallate, lauryl gallate, LG
- (23) 2,4,5-trihydroxy-butyrophenone = THBP
- (24) 2,2'-methylene-bis-(4-ethyl-6-tert.-butyl-phenol)
- 5 (25) 1,1-bis-(2-hydroxy-3,5-dimethyl-phenyl)-butane
- (26) 1,1'-methylene-bis-(naphthol-2)
- (27) 2,2-bis-(4-hydroxy-phenyl)-propane = bisphenol A
- (28) mixture of tert.-butylated 2,2-bis-(4-hydroxy-phenyl)-propanes
- (29) bis-3,3-bis-(4-hydroxy-3-tert.-butyl-phenyl)-butanoic acid glycol ester =
- 10 DTB glycol ester
- (30) 1,1-bis-(5-tert.-butyl-4-hydroxy-2-methyl-phenyl)-butane
- (31) 1,1,3-tris-(5-tert.-butyl-4-hydroxy-2-methyl-phenyl)-butane
- (32) 4,4'-methylene-bis-(2-tert.-butyl-6-methyl-phenol)
- (33) 4,4'-methylene-bis-(2,6-di-tert.-butyl-phenol)
- 15 (34) 4,4'-methylene-bis-(2,5-di-tert.-butyl-phenol)
- (35) 1,1-bis-(4-hydroxy-phenyl)-cyclohexane
- (36) 1,1-bis-(3-cyclohexyl-4-hydroxy-phenyl)-cyclohexane
- (37) 1,3,5-trimethyl-2,4,6-tris-(3,5-di-tert.-butyl-4-hydroxy-benzyl)-benzene:



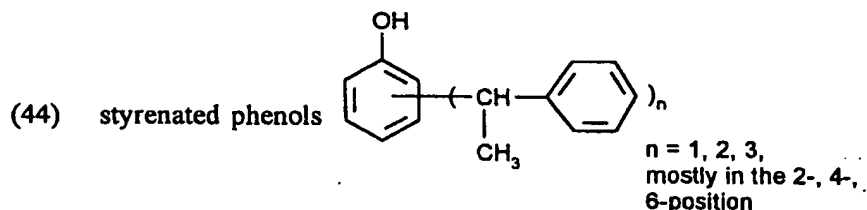
- 20 (38) catechol
- (39) 4-tert.-butyl-catechol = TBC; 1,2-dihydroxy-4-tert.-butylbenzene
- (40) hydroquinone
- (41) 4-methoxyphenol, hydroquinone monomethyl ether
- (42) 4-benzyloxyphenol, hydroquinone monobenzyl ether
- 25 (43) mixtures of 3-tert.-butyl-4-hydroxy-anisole



3-isomer



2-isomer



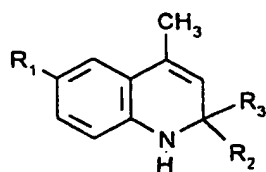
- (45) 3,5-di-tert.-butyl-4-hydroxy-benzyl alcohol
- (46) 2,6-di-tert.-butyl-4-methoxy-phenol
- 5 (47) octadecyl-3-(3,5-di-tert.-butyl-4-hydroxy-phenyl)-propionate
- (48) pentaerythrityl-tetrakis-[3-(3,5-di-tert.-butyl-4-hydroxy-phenyl)-propionate]
- (49) 1,6-bis-[3-(3,5-di-tert.-butyl-4-hydroxy-phenyl)-propionyloxy]-n-hexane
- (50) 2,2-bis-(3,5-di-tert.-butyl-4-hydroxy-benzyl)-malonic acid-di n-octyl ester
- (51) 2,2'-methylene-bis-(4,6-dimethyl-phenol)
- 10 (52) 2,2'-methylene-bis-(6-tert.-butyl-4-methyl-phenol)
- $R = -C(CH_3)_3$
- (53) 2,2'-methylene-bis-(4-methyl-6-nonyl-phenol)
- (54) 2,2'-methylene-bis-[4-methyl-6-(1-methyl-cyclohexyl)-phenol]
- (55) 2,2'-methylene-bis[4-methyl-(6- α -methyl-benzyl)-phenol]

15 **Table 3:**

- (1) N,N'-di-sec.-butyl-p-phenylenediamine = DBPPD
- (2) N,N'-bis-(1,4-dimethyl-pentyl)-p-phenylenediamine
- (3) N,N'-bis-(1-ethyl-3-methyl-pentyl)-p-phenylenediamine
- (4) N,N'-bis-(1-methyl-heptyl)-p-phenylenediamine
- 20 (5) N,N'-dicyclohexyl-p-phenylenediamine
- (6) N,N'-diphenyl-p-phenylenediamine = DPPD
- (7) N,N'-di-(naphthyl-2)-p-phenylenediamine
- (8) N-isopropyl-N'-phenyl-p-phenylenediamine
- (9) N-(1,3-dimethyl-butyl)-N'-phenyl-p-phenylenediamine

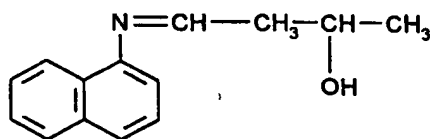
- (10) N-(1-methyl-heptyl)-N'-phenyl-p-phenylenediamine
 (11) N-cyclohexyl-N'-phenyl-p-phenylenediamine
 (12) 4-(p-toluene-sulphonamido)-diphenylamine
 (13) N,N'-dimethyl-N,N'-di-sec.-butyl-p-phenylenediamine
 5 (14) diphenylamine
 (15) 4-isopropoxy-diphenylamine
 (16) N-phenyl-1-naphthylamine
 (17) N-phenyl-2-naphthylamine
 (18) octylated diphenylamine, predominantly 4-octyl-diphenylamine
 10 (19) 4-n-butylamino-phenol
 (20) 4-butyrylamino-phenol
 (21) 4-nonanoylamino-phenol
 (22) 4-dodecanoylamino-phenol
 (23) 4-octadecanoyl-amino-phenol
 15 (24) di-(4-methoxy-phenyl)-amine
 (25) 2,6-di-tert.-butyl-4-dimethylamino-methyl-phenol
 (26) 2,4'-diamino-diphenylmethane
 (27) 4,4'-diamino-diphenylmethane
 (28) N,N,N',N'-tetramethyl-4,4'-diamino-diphenylmethane
 20 (29) 1,2-di-(phenylamino)-ethane
 (30) 1,2-di-[(2-methyl-phenyl)-amino]-ethane
 (31) 1,3-di-(phenylamino)-propane
 (32) (o-tolyl)-biguanide
 (33) the condensation product of aniline and acetaldehyde,
 25 (34) aniline-aldol-condensate
 (35) the product from aniline and butyraldehyde
 (36) (polymeric) 2,2,4-trimethyl-1,2-dihydroquinoline
 (37) aniline-acetone condensate
 (38) 6-ethoxy-2,2,4-trimethyl-1,3-dihydroquinoline = ethoxyquin

30

 $R_1 = -OC_2H_5$ $R_2 = -CH_3$

- (39) 6-dodecyl-2,2,4-trimethyl-1,2-dihydroquinoline
 (40) 2,2,4-trimethyl-6-phenyl-1,2-dihydroquinoline

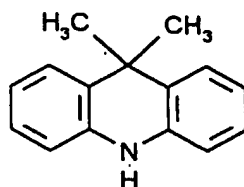
(41) 1-aminonaphthalene-aldol condensate



(42) the product of 2-phenylamino-naphthalene and acetone

(43) diphenylamine-acetone condensate, compounds of 5,5-dimethyl-acridine, etc.

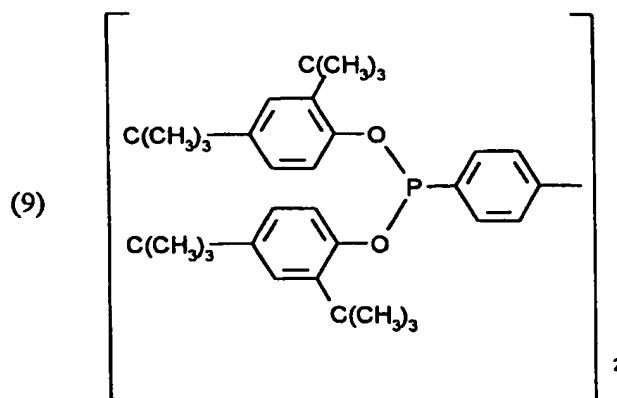
5



(44) N,N'-diisopropyl-p-phenylenediamine

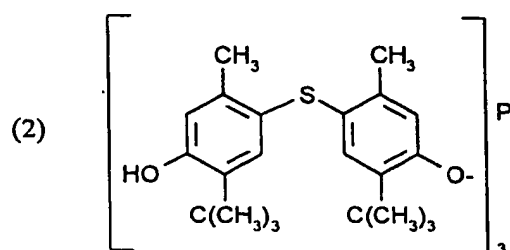
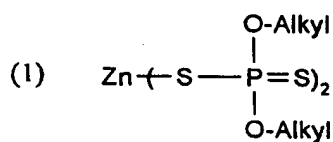
Table 5:

- | | | |
|----|-----|-------------------------------------|
| | (1) | triphenylphosphine |
| 10 | (2) | diethyl phosphite |
| | (3) | triphenyl phosphite |
| | (4) | tris-nonylphenyl phosphite |
| | (5) | tris-(mono-dinonylphenyl)-phosphite |
| | (6) | tridecyl phosphite |
| 15 | (7) | tri-isodecyl phosphite |
| | (8) | tri-dodecyl phosphite |

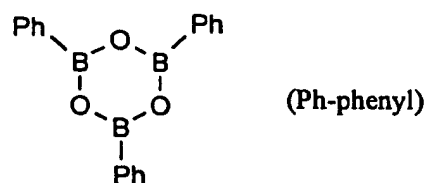
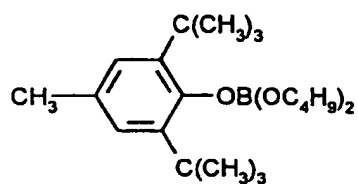
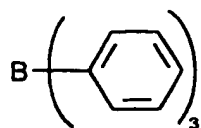
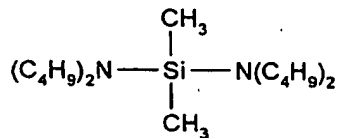
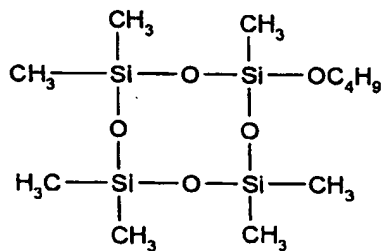


- (10) the condensation product of 4,4'-thio-bis-(2-tert.-butyl-5-methyl-phenol)
- (11) octyl-diphenyl phosphite
- (12) isooctyl-diphenyl phosphite
- 5 (13) decyl-diphenyl phosphite
- (14) isodecyl-diphenyl phosphite
- (15) didecyl-phenyl phosphite
- (16) diisodecyl-phenyl phosphite
- (17) 3,5-di-tert.-butyl-4-hydroxybenzyl-phosphonic acid diethyl ester
- 10 (18) 3,5-di-tert.-butyl-4-hydroxybenzyl-phosphonic acid di-n-octadecyl ester

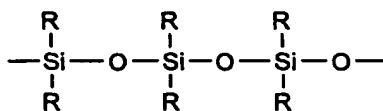
Zinc, barium and calcium thiophosphates, e.g. of formula




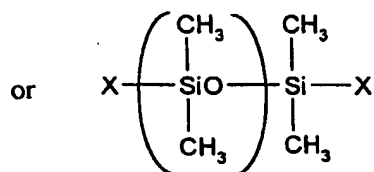
boric acid esters of formula


$$\begin{array}{c} \text{C}_4\text{H}_9 \quad \text{O} \\ \diagup \quad \parallel \\ \text{Si} \quad \text{O}-\text{C}-(\text{CH}_2)_2 \\ \diagdown \quad \parallel \\ \text{C}_4\text{H}_9 \quad \text{O} \end{array}$$


organopolysiloxanes of formula



where R = -CH₃, -Ph, -H or $-(\text{CH}_2)_3\text{O}-\text{CH}_2-\text{CH}-\text{CH}_2$ 



where n = 1-10 and

5 X = $-\text{CH}_2\text{O}-(\text{CH}_2)_3-\text{O}(\text{CH}_2)_3-\text{OH}$
 $-(\text{CH}_2)_4-\text{OH}$
 $-\text{OC}_2\text{H}_5,$

or sterically hindered based on 2,6-tetramethyl-piperidine.